

Revisit of the Orbital-Fluctuation-Mediated Superconductivity in LiFeAs: Nontrivial Spin-Orbit Interaction Effects on the Bandstructure and Superconducting Gap Function

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Precise gap structure in LiFeAs ($T_c = 18\text{K}$) given by ARPES studies offers us significant information to understand the pairing mechanism in iron-based superconductors. The most remarkable characteristics in LiFeAs gap structure would be that “the largest gap emerges on the tiny hole-pockets around Z point”. This result had been naturally explained in terms of the orbital-fluctuation scenario (T. Saito *et al.*, Phys. Rev. B **90**, 035104 (2014)), whereas an opposite result is obtained by the spin-fluctuation scenario. In this paper, we study the gap structure in LiFeAs by taking the spin-orbit interaction (SOI) into account, motivated by the recent ARPES studies that revealed the significant SOI-induced modification of the Fermi surface topology. For this purpose, we construct the two possible tight-binding models with finite SOI by referring the bandstructures given by different ARPES groups. In addition, we extend the gap equation for multiorbital systems with finite SOI, and calculate the gap functions by applying the orbital-spin fluctuation theory. On the basis of both SOI-induced band structures, main characteristics of the gap structure in LiFeAs are naturally reproduced only in the presence of strong inter-orbital interactions between ($d_{xz/yz}$ - d_{xy}) orbitals. Thus, the experimental gap structure in LiFeAs is a strong evidence for the orbital-fluctuation pairing mechanism.

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I. INTRODUCTION

The pairing mechanism of iron-based superconductors had been studied intensively as a central issue. Up to now, the spin-fluctuation-mediated s_{\pm} -wave state with sign reversal [1–5] and orbital-fluctuation-mediated s_{++} -wave state without sign reversal [6–8] had been proposed. According to theoretical studies for realistic multiorbital models, s_{\pm} -wave state is fragile against impurities: s_{\pm} -wave state with $T_c = 30\text{K}$ disappears when the residual resistivity ρ_0 is as large as $\sim 15 \mu\Omega\text{cm}$ for $m^*/m_b \sim 3$ by $3d$ -transition metal impurities [9, 10]. Experimentally, the superconductivity in LaFeAsO+F, (K,Ba)Fe₂As₂ and Ba(Fe,Co)₂As₂ survives till $\rho_0 \sim 300 \mu\Omega\text{cm}$ [11–14], indicate that the s_{++} -wave state occurs in these compounds. On the other hand, $T_c = 18\text{K}$ disappears only when $\rho_0 \sim 40 \mu\Omega\text{cm}$ in Ba(Fe,Ru)₂As₂ by electron irradiation measurement [15]. In the neutron spectroscopy, the resonance-like peaks in various compounds are also explained in terms of the s_{++} -wave state by taking the realistic quasiparticle inelastic scattering into account [16].

In many Fe-based superconductors, the gap structure has been determined in detail by the angle-resolved photoemission spectroscopy (ARPES) studies. Although ARPES is not a sign-sensitive experiment, the detailed gap structure given by the ARPES offers us very useful information to distinguish the pairing mechanism. In fact, orbital dependence of the gap function is very crucial since it reflects the orbital dependence of the pairing interaction strength. In the spin-fluctuation mechanism, the superconductivity is mainly induced by the intra-orbital scattering of the Cooper-pairs, since the spin-

fluctuations are driven by the intra-orbital Coulomb interaction U . For this reason, the gap magnitude $|\Delta(\mathbf{k})|$ tends to largely depend on the orbital character of the Fermi surface in the spin-fluctuation mechanism. In contrast, in the orbital-fluctuation mechanism, the superconductivity is induced by the inter-orbital scattering of the Cooper-pairs. For this reason, the orbital dependence of $|\Delta(\mathbf{k})|$ tends to be moderate in the orbital-fluctuation mechanism.

Motivated by the above discussion, the gap magnitude on the d_{z^2} -orbital hole-pocket around Z point in Ba122 and Sr122 compounds had been studied intensively. Because of the absence of the intra- d_{z^2} -orbital nesting, the spin-fluctuation scenario predicts the emergence of the “horizontal node” on the hole-pocket around Z point [17, 18]. In contrast, the horizontal node is absent in the orbital-fluctuation-mediated s -wave state, since the d_{z^2} -orbital electrons contribute to the orbital-fluctuations [18]. In BaFe₂(As,P)₂ and Ba(Fe,Co)₂As₂, several ARPES groups reported the absence of the horizontal node [19–21], whereas horizontal node was reported in Ref. [22].

For studying the pairing mechanism, LiFeAs is an suitable compound since the detailed band structure and the superconducting gap functions had been determined by several ARPES groups [23, 24]. LiFeAs is an ideal system for ARPES studies in that the charge neutral cleavage plane exists and clean single crystals can be synthesized. Therefore, the intrinsic gap structure free from extrinsic effects (such as the impurity effect) is obtained by ARPES studies. The experimental band structure and Fermi surfaces (FSs) are shown in Fig. 1: Reflect-

ing the bad nesting between the hole-like FSs (h-FSs) and electron-like FSs (e-FSs) in LiFeAs, moderate spin-fluctuations are observed by NMR studies [25] and neutron scattering studies [26–28]. Interestingly, in LiFeAs, the largest gap appears in the tiny three-dimensional (3D) hole-pockets (h-FS1,2) according to the ARPES studies [23, 24] and the Scanning Tunneling Microscopy (STM) study [29].

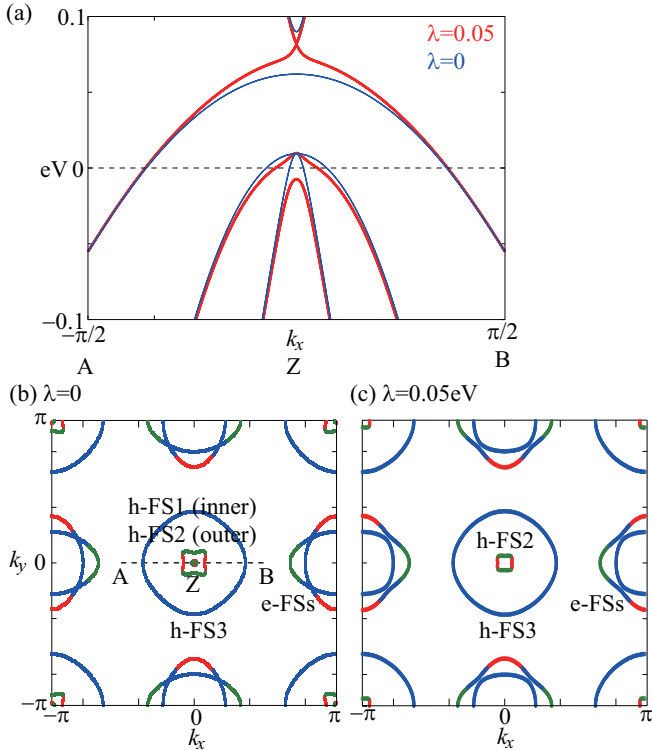


FIG. 1: (Color online) (a) The dispersion of the band structure between points A and B for $\lambda = 0.05$ eV (red lines) and 0 (blue lines), respectively. We call the former the “SOI-induced bandstructure (I)”. A and B points are shown in (b). The corresponding FSs for (b) $\lambda = 0$ and (c) 0.05 eV in the $k_z = \pi$ plane are shown respectively. In (c), h-FS1 disappears due to the SOI. Green, red, and blue lines correspond to xz , yz , and xy orbitals, respectively.

Based on the tight-binding model for LiFeAs constructed from the ARPES band structure, some authors studied the possible pairing scenarios. In the spin-fluctuation mechanism for the s_{\pm} -wave state, the gaps on the tiny hole-pockets, h-FS1,2 in Fig. 1 (b), are very small, because of the bad intra-orbital nesting between h-FS1,2 and e-FSs [31, 32]. In contrast, in the orbital-fluctuation mechanism for the s_{++} -wave state, the gaps on the tiny hole-pockets are the largest, because of the good inter-orbital nesting [32]. The latter theoretical result is consistent with experiments [23, 24, 29].

However, although the SOI had been neglected in previous theoretical studies in Refs. [31, 32], recent ARPES studies for LiFeAs revealed that the SOI is never negligible to construct the FSs and the band structure near

the Fermi level. For instance, the degeneracy of the $d_{xz/yz}$ hole bands on the Γ -Z line is lifted by the SOI [33–35]. For this reason, one of the tiny two hole-pockets (h-FS1,2) disappears due to the SOI, as shown in Fig. 1 (c) (or Fig. 6 (d)). Similar disappearance of h-FS1 due to SOI had been observed in FeSe [36–41]. Since the tiny hole-pockets play important roles in the pairing mechanism, a theoretical study for LiFeAs including the SOI had been highly required.

In this paper, we study the gap structure of LiFeAs by taking the effect of the SOI into account, in order to distinguish the correct pairing scenario. For this purpose, we construct two possible 3D ten-orbital models, which correspond to the different experimental band structures reported in Refs. [33, 34]. Both models possess a single tiny hole-pocket around Z point. We also introduce both the multi-orbital Coulomb interaction (U, U', J) and the quadrupole interaction (g): The latter will originate from the Aslamazov-Larkin type vertex correction (AL-VC), which describes the many-body effect beyond the mean-field approximation [7, 8, 42, 43]. By using the random-phase-approximation (RPA), the orbital- and spin-fluctuations develop with increasing g and U , respectively. Then, we extend the gap equation for multi-orbital systems with finite SOI, and calculate the gap function by using the pairing interaction given by the RPA. In the s_{++} -wave state due to orbital-fluctuations, the gap on the smallest h-FS is the largest of all. In contrast, in the s_{\pm} -wave state due to spin-fluctuations, the gap on the smallest h-FS is almost zero. Therefore, the superconductivity in LiFeAs originates from the inter-orbital ($d_{xz/yz}$ - d_{xy}) interactions due to orbital-fluctuations, not from the intra-orbital spin-fluctuation interactions.

When both the orbital- and spin-fluctuations develop comparably, we frequently obtain the “hole- s_{\pm} -state”, in which the gap structure has the “sign reversal between h-FSs”. This interesting gap structure had been discussed in (Ba,K)Fe₂As₂ experimentally [44, 45]. In Appendix A, we show that the hole- s_{\pm} -wave gap structure can be obtained in the 3D model for Ba122 system.

II. FORMALISM

A. SOI-induced bandstructure (I) and interaction terms

In this paper, we set x and y axes parallel to the nearest Fe-Fe bonds, and the orbital z^2 , xz , yz , xy , and $x^2 - y^2$ of Fe-A (B) site are denoted as 1, 2, 3, 4, and 5 (6, 7, 8, 9, and 10) respectively. We used the 3D ten-orbital tight-binding model which is given in Ref. [31]. This model is obtained by fitting the experimentally observed dispersion reported in Ref. [23]. Note that the band renormalization due to the mass enhancement $m^*/m_b = 2 \sim 3$ is taken into account in the present model.

We stress that the experimental FSs of LiFeAs in Fig. 1, on which the present study is based, are very differ-

ent from the FSs given by the density functional theory (DFT), in which h-FS1,2 predicted by the DFT are much larger. Better agreement between theory and ARPES is achieved by the LDA+DMFT study [30], since the FS1,2 shrinks due to the orbital-dependent self-energy that is absent in the LDA.

In the absence of the SOI, the kinetic term of the ten-orbital model is given as

$$\begin{aligned}\hat{H}^0 &= \sum_{ablm\sigma} t_{l,m}(\mathbf{R}_a - \mathbf{R}_b) c_{l\sigma}^\dagger(\mathbf{R}_a) c_{m\sigma}(\mathbf{R}_b) \\ &= \sum_{\mathbf{k}lm\sigma} \left\{ \sum_a t_{l,m}(\tilde{\mathbf{R}}_a) e^{i\mathbf{k} \cdot \tilde{\mathbf{R}}_a} \right\} c_{l\sigma}^\dagger(\mathbf{k}) c_{m\sigma}(\mathbf{k}) \quad (1)\end{aligned}$$

where $l, m = 1 - 5$ ($6 - 10$) represent the d orbitals at Fe-A (Fe-B) atom, and $\sigma = \pm 1$ is the spin index. $\tilde{\mathbf{R}}_a$ is the Fe-site position measured from an Fe-A atom, $c_{l\sigma}^\dagger(\mathbf{k})$ is the creation operator of the d electron, and $t_{l,m}(\tilde{\mathbf{R}})$ is the hopping integral. The values of $t_{l,m}(\tilde{\mathbf{R}})$ are shown in Ref. [31].

Next, we introduce the interaction terms. We consider both the multiorbital Coulomb interaction ($U, U', J = (U - U')/2$) and the quadrupole interaction. The quadrupole interaction Hamiltonian is given as

$$V_{\text{quad}} = -g(\omega_l) \sum_a^{\text{site}} \left(\hat{O}_{yz}^a \cdot \hat{O}_{yz}^a + \hat{O}_{xz}^a \cdot \hat{O}_{xz}^a \right) \quad (2)$$

where $g(\omega_l) = g\omega_0^2/(\omega_l^2 + \omega_0^2)$, and $g = g(0)$ is the quadrupole interaction at $\omega_l = 0$. ω_0 is the cutoff energy of the quadrupole interaction. $\hat{O}_\Gamma^a = \sum_{l,m} o_\Gamma^{l,m} \hat{m}_{l,m}^a$ ($\hat{m}_{l,m}^a = \sum_\sigma c_{l\sigma}^\dagger(\mathbf{R}_a) c_{m\sigma}(\mathbf{R}_a)$) is the quadrupole operator at site \mathbf{R}_a introduced in Ref. [6]: The non-zero coefficients of $o_\Gamma^{l,m} = o_\Gamma^{m,l}$ are $o_{xz}^{2,5} = o_{xz}^{3,4} = \sqrt{3}o_{xz}^{1,2} = 1$, and $-o_{yz}^{3,5} = o_{yz}^{2,4} = \sqrt{3}o_{yz}^{1,3} = 1$. Thus, \hat{V}_{quad} has many non-zero inter-orbital elements. As explained in Ref. [6], g is given by in-plane Fe-ion oscillations. Also, the AL-VC due to Coulomb interaction gives large effective quadrupole interaction g [7].

Now, we analyze the present model by applying the RPA. The $32 \times 32 \times 16$ \mathbf{k} meshes are used in the numerical study. We fix the temperature at $T = 0.01$ eV, the cutoff energy at $\omega_0 = 0.02$ eV, and set the filling of each Fe-site as $n = 6.0$. The irreducible susceptibility is given as

$$\chi_{ll',mm'}^0(q) = -\frac{T}{N} \sum_{\mathbf{k}} G_{l,m}(\mathbf{k} + q) G_{m',l'}(\mathbf{k}), \quad (3)$$

where $q = (\mathbf{q}, \omega_l)$ and $\mathbf{k} = (\mathbf{k}, \epsilon_n)$. $\epsilon_n = (2n + 1)\pi T$ and $\omega_l = 2l\pi T$ are the fermion and boson Matsubara frequencies. $\hat{G}(\mathbf{k}) = [i\epsilon_n + \mu - \hat{h}^0(\mathbf{k})]^{-1}$ is the Green function in the orbital representation, where $\hat{h}^0(\mathbf{k})$ is the matrix representation of \hat{H}^0 for the momentum \mathbf{k} , and μ is the chemical potential. In the RPA, the spin and charge susceptibilities in the matrix form are given by

[46]

$$\hat{\chi}^s(q) = \frac{\hat{\chi}^0(q)}{\hat{1} - \hat{\Gamma}^s \hat{\chi}^0(q)}, \quad (4)$$

$$\hat{\chi}^c(q) = \frac{\hat{\chi}^0(q)}{\hat{1} - \hat{\Gamma}^c(\omega_l) \hat{\chi}^0(q)}, \quad (5)$$

where

$$(\Gamma^s)_{l_1 l_2, l_3 l_4}^{\alpha\beta} = \delta_{\alpha\beta} \times \begin{cases} U, & l_1 = l_2 = l_3 = l_4 \\ U', & l_1 = l_3 \neq l_2 = l_4 \\ J, & l_1 = l_2 \neq l_3 = l_4 \\ J', & l_1 = l_4 \neq l_2 = l_3 \\ 0, & \text{otherwise} \end{cases} \quad (6)$$

$$\hat{\Gamma}^c(\omega_l) = -\hat{C} - 2\hat{V}_{\text{quad}}(\omega_l), \quad (7)$$

$$(C)_{l_1 l_2, l_3 l_4}^{\alpha\beta} = \delta_{\alpha\beta} \times \begin{cases} U, & l_1 = l_2 = l_3 = l_4 \\ -U' + 2J, & l_1 = l_3 \neq l_2 = l_4 \\ 2U' - J, & l_1 = l_2 \neq l_3 = l_4 \\ J', & l_1 = l_4 \neq l_2 = l_3 \\ 0, & \text{otherwise} \end{cases} \quad (8)$$

$\alpha, \beta (= A, B)$ is Fe-site. In this paper, we set $U = U' + 2J$, $J = J'$, and $J = U/6$.

In the RPA, the enhancement of the spin susceptibility $\hat{\chi}^s$ originates from the intra-orbital Coulomb interaction U and the “intra-orbital nesting” of the FSs. On the other hand, the enhancement of $\hat{\chi}^c$ originates from the quadrupole-quadrupole interaction in Eq. (2) and the “inter-orbital nesting” of the FSs. The magnetic (orbital) order occurs when $\alpha_{s(c)} = 1$, where $\alpha_{s(c)}$ is the maximum eigenvalue of $\hat{\Gamma}^{s(c)} \hat{\chi}^{(0)}(\mathbf{q}, 0)$, called the spin (charge) Stoner factor. Here, the magnetic order is realized when U reaches $U_{\text{cr}} = 0.448$ eV. Also, the orbital order is realized when g reaches $g_{\text{cr}} = 0.132$ eV for $U = 0$ [32]. The used interactions are smaller since the energy-scale had been renormalized by $z = (m^*/m_b)^{-1}$ in the present tight-binding model.

B. Spin-orbit interaction

In this subsection, we introduce the SOI. In the presence of SOI for d electron, the 20×20 matrix expression of the total Hamiltonian at \mathbf{k} is given by

$$\begin{aligned}\hat{h}(\mathbf{k}) &= \begin{pmatrix} \hat{h}^0(\mathbf{k}) + \lambda \hat{l}_z/2 & \lambda(\hat{l}_x - i\hat{l}_y)/2 \\ \lambda(\hat{l}_x + i\hat{l}_y)/2 & \hat{h}^0(\mathbf{k}) - \lambda \hat{l}_z/2 \end{pmatrix} \\ &\equiv \begin{pmatrix} \hat{h}^{\uparrow\uparrow}(\mathbf{k}) & \hat{h}^{\uparrow\downarrow}(\mathbf{k}) \\ \hat{h}^{\downarrow\uparrow}(\mathbf{k}) & \hat{h}^{\downarrow\downarrow}(\mathbf{k}) \end{pmatrix}, \quad (9)\end{aligned}$$

where the first and the second rows (columns) correspond to \uparrow -spin and \downarrow -spin. λ is coupling constant of SOI. The

matrix elements for \hat{l} for d -orbital, which is diagonal with respect to site, are given by [47]

$$\hat{l}_x = \begin{pmatrix} 0 & 0 & \sqrt{3}i & 0 & 0 \\ 0 & 0 & 0 & i & 0 \\ -\sqrt{3}i & 0 & 0 & 0 & -i \\ 0 & -i & 0 & 0 & 0 \\ 0 & 0 & i & 0 & 0 \end{pmatrix}, \quad (10)$$

$$\hat{l}_y = \begin{pmatrix} 0 & -\sqrt{3}i & 0 & 0 & 0 \\ \sqrt{3}i & 0 & 0 & 0 & -i \\ 0 & 0 & 0 & -i & 0 \\ 0 & 0 & i & 0 & 0 \\ 0 & i & 0 & 0 & 0 \end{pmatrix}, \quad (11)$$

$$\hat{l}_z = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -i & 0 & 0 \\ 0 & i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2i \\ 0 & 0 & 0 & -2i & 0 \end{pmatrix}, \quad (12)$$

where the first to fifth rows (columns) correspond to d orbitals $z^2, xz, yz, xy, x^2 - y^2$, respectively.

The red (blue) lines in Fig. 1 (a) shows the band structure of LiFeAs for $\lambda = 0.05$ eV ($\lambda = 0$), respectively. Points A and B are shown in Fig. 1 (b). When $\lambda = 0$, the two bands composed of $d_{xz} + d_{yz}$ orbital near the Fermi level at Z point are degenerated, however, this degeneracy is lifted by the SOI, which seems to be observed by recent ARPES measurements in LiFeAs [33, 34] as well as in FeSe [36–41]. Figures 1 (b) and (c) show the FSs of LiFeAs for $\lambda = 0$ and 0.05 eV, respectively. For this reason, h-FS1 disappears by introducing the SOI. This model is consistent with the ARPES measurement by Miao *et al.* in the way that only outer band of two $d_{xz/yz}$ -orbital hole band at Z point crosses the Fermi level [34]. In Sec. IV, we also construct the tight-binding model that gives the ARPES band structure reported by Borisenko *et al* [33].

Since the energy-scale of the present tight-binding model had been renormalized by the factor $z = (m^*/m_b)^{-1}$, we should use a renormalized λ in Eq. (9) in principle. However, we hereafter use a typical λ for Fe ion ($\lambda = 0.05$ eV) [47] without considering the renormalization, in order to clarify the effect of the SOI. In fact, we had verified that the obtained gap structures in later sections are unchanged by using smaller SOI coupling, say $\lambda \sim 0.02$ eV.

C. Gap equation

In this subsection, we introduce the linearized gap equation in the presence of the SOI. In the 3D model, it is difficult to analyze fine momentum dependence of the SC gap on the FSs by using a conventional 3D \mathbf{k} -meshes. In this paper, we used 48×16 \mathbf{k} points for each FS sheet in solving the gap equation in order to obtain the precise gap structure [48]. In the absence of impurities, the linearized gap equation for the singlet state is

given as

$$\lambda_E \Delta_{\Sigma\bar{\Sigma}}^L(\mathbf{k}, \epsilon_n) = \frac{\pi T}{(2\pi)^3} \sum_{\epsilon_m} \sum_M \sum_{\Lambda}^{\text{FS}} \int_{\text{FSM}}^{\uparrow\downarrow} \frac{d\mathbf{k}'_{\text{FSM}}}{v^M(\mathbf{k}')} \times V_{\Sigma\bar{\Sigma}\Lambda\bar{\Lambda}}^{LM}(\mathbf{k}, \mathbf{k}', \epsilon_n - \epsilon_m) \frac{\Delta_{\Lambda\bar{\Lambda}}^M(\mathbf{k}', \epsilon_m)}{|\epsilon_m|}, \quad (13)$$

where λ_E is the eigenvalue of the gap equation. λ_E reaches unity at $T = T_c$. We solve the gap function for the largest eigenvalue, since it corresponds to the pairing state for the highest T_c . L and M denote the FSs, and Σ and Λ denote the pseudo-spin (\uparrow, \downarrow) in the presence of the SOI. Note that $\bar{\Sigma} \equiv -\Sigma$. $\Delta_{\Sigma\bar{\Sigma}}^L(\mathbf{k}, \epsilon_n)$ is the pseudo-spin singlet gap function on the L -th FS. In Eq. (13), we perform the surface integral on the M -th FS. The pairing interaction V in Eq. (13) for the pseudo-spin singlet pair is given by

$$V_{\Sigma\bar{\Sigma}\Lambda\bar{\Lambda}}^{LM}(\mathbf{k}, \mathbf{k}', \epsilon_n - \epsilon_m) = \sum_{lmm'l'} \sum_{\sigma\lambda\lambda'\sigma'} U_{lL}^{\sigma\Sigma*}(\mathbf{k}) U_{l'L'}^{\sigma'\bar{\Sigma}*}(-\mathbf{k}) \times V_{lmm'l'}^{\sigma\lambda\lambda'\sigma'}(\mathbf{k} - \mathbf{k}', \epsilon_n - \epsilon_m) \times U_{m'M'}^{\lambda'\bar{\Lambda}}(-\mathbf{k}') U_{mM}^{\lambda\Lambda}(\mathbf{k}'), \quad (14)$$

where σ, λ mean real spin (\downarrow, \uparrow) and $U_{lL}^{\sigma\Sigma}(\mathbf{k}) = \langle \mathbf{k}; l\sigma | \mathbf{k}; L\Sigma \rangle$ is the unitary matrix connecting between the band representation and the orbital one. Here, the pairing interaction in the orbital basis is given by

$$V_{lmm'l'}^{\sigma\lambda\lambda'\sigma'} = V_{lmm'l'}^c \delta_{\sigma\lambda} \delta_{\sigma'\lambda'} + V_{lmm'l'}^s \boldsymbol{\sigma}_{\sigma\lambda} \cdot \boldsymbol{\sigma}_{\sigma'\lambda'} = \begin{cases} V_{lmm'l'}^c + V_{lmm'l'}^s, & \sigma = \lambda = \lambda' = \sigma', \\ V_{lmm'l'}^c - V_{lmm'l'}^s, & \sigma = \lambda \neq \lambda' = \sigma', \\ 2V_{lmm'l'}^s, & \sigma = \lambda' \neq \lambda = \sigma', \\ 0, & \text{otherwise,} \end{cases} \quad (15)$$

$$\hat{V}^c = \frac{1}{2} \hat{\Gamma}^c \hat{\chi}^c \hat{\Gamma}^c, \quad \hat{V}^s = \frac{1}{2} \hat{\Gamma}^s \hat{\chi}^s \hat{\Gamma}^s. \quad (16)$$

In this paper, $\hat{V}^\xi (\xi = c, s)$ is calculated without the SOI, since it is verified that the SOI is negligible except at the vicinity of the magnetic critical point ($\alpha_s \approx 1$).

Now, we introduce the relation between $U_{lL}^{\sigma\Sigma}(\mathbf{k})$ and $U_{lL}^{\sigma\bar{\Sigma}}(-\mathbf{k})$ to preserve the time-reversal symmetry between the states $|\mathbf{k}; L\Sigma\rangle$ and $|\mathbf{k}; L\bar{\Sigma}\rangle$. The 20×20 Hamiltonian $\hat{h}(\mathbf{k})$ is given in Eq. (9). Then, $\hat{h}(-\mathbf{k})^*$ is given by

$$\begin{aligned} \hat{h}(-\mathbf{k})^* &= \begin{pmatrix} \hat{h}_0(-\mathbf{k})^* + \lambda \hat{l}_z^*/2 & \lambda(\hat{l}_x - i\hat{l}_y)^*/2 \\ \lambda(\hat{l}_x + i\hat{l}_y)^*/2 & \hat{h}_0(-\mathbf{k})^* - \lambda \hat{l}_z^*/2 \end{pmatrix} \\ &= \begin{pmatrix} \hat{h}_0(\mathbf{k}) - \lambda \hat{l}_z/2 & -\lambda(\hat{l}_x + i\hat{l}_y)/2 \\ -\lambda(\hat{l}_x - i\hat{l}_y)/2 & \hat{h}_0(\mathbf{k}) + \lambda \hat{l}_z/2 \end{pmatrix} \\ &= \begin{pmatrix} \hat{h}^{\downarrow\downarrow}(\mathbf{k}) & -\hat{h}^{\downarrow\uparrow}(\mathbf{k}) \\ -\hat{h}^{\uparrow\downarrow}(\mathbf{k}) & \hat{h}^{\uparrow\uparrow}(\mathbf{k}) \end{pmatrix}. \end{aligned} \quad (17)$$

Here, we express the unitary matrix as $\hat{U}(\mathbf{k}) \equiv \begin{pmatrix} \hat{U}^{\uparrow\uparrow}(\mathbf{k}) & \hat{U}^{\uparrow\downarrow}(\mathbf{k}) \\ \hat{U}^{\downarrow\uparrow}(\mathbf{k}) & \hat{U}^{\downarrow\downarrow}(\mathbf{k}) \end{pmatrix}$ that diagonalize $\hat{h}(\mathbf{k})$. That is,

$$\hat{U}(\mathbf{k})^\dagger \hat{h}(\mathbf{k}) \hat{U}(\mathbf{k}) = \begin{pmatrix} \hat{E}(\mathbf{k}) & \hat{0} \\ \hat{0} & \hat{E}(\mathbf{k}) \end{pmatrix}, \quad (18)$$

where $\hat{E}(\mathbf{k})$ is diagonal. By using Eqs. (17) and (18), we obtain that

$$\begin{pmatrix} \hat{U}^{\downarrow\downarrow}(\mathbf{k})^\dagger & -\hat{U}^{\uparrow\downarrow}(\mathbf{k})^\dagger \\ -\hat{U}^{\downarrow\uparrow}(\mathbf{k})^\dagger & \hat{U}^{\uparrow\uparrow}(\mathbf{k})^\dagger \end{pmatrix} \hat{h}(-\mathbf{k})^* \times \begin{pmatrix} \hat{U}^{\downarrow\downarrow}(\mathbf{k}) & -\hat{U}^{\downarrow\uparrow}(\mathbf{k}) \\ -\hat{U}^{\uparrow\downarrow}(\mathbf{k}) & \hat{U}^{\uparrow\uparrow}(\mathbf{k}) \end{pmatrix} = \begin{pmatrix} \hat{E}(\mathbf{k}) & \hat{0} \\ \hat{0} & \hat{E}(\mathbf{k}) \end{pmatrix}. \quad (19)$$

Therefore, $\hat{U}(-\mathbf{k})$ is related to $\hat{U}(\mathbf{k})$ as

$$\hat{U}(-\mathbf{k}) = \begin{pmatrix} \hat{U}^{\downarrow\downarrow}(\mathbf{k})^* & -\hat{U}^{\downarrow\uparrow}(\mathbf{k})^* \\ -\hat{U}^{\uparrow\downarrow}(\mathbf{k})^* & \hat{U}^{\uparrow\uparrow}(\mathbf{k})^* \end{pmatrix}. \quad (20)$$

That is, the relations between $U_{iL}^{\sigma\Sigma}(\mathbf{k})$ and $U_{iL}^{\sigma\bar{\Sigma}}(-\mathbf{k})$ are summarized as

$$\begin{aligned} U_{iL}^{\uparrow\uparrow}(-\mathbf{k}) &= U_{iL}^{\downarrow\downarrow}(\mathbf{k})^*, \\ U_{iL}^{\downarrow\downarrow}(-\mathbf{k}) &= U_{iL}^{\uparrow\uparrow}(\mathbf{k})^*, \\ U_{iL}^{\uparrow\downarrow}(-\mathbf{k}) &= -U_{iL}^{\downarrow\uparrow}(\mathbf{k})^*, \\ U_{iL}^{\downarrow\uparrow}(-\mathbf{k}) &= -U_{iL}^{\uparrow\downarrow}(\mathbf{k})^*. \end{aligned} \quad (21)$$

These relations are necessary to obtain physical gap functions.

Next, we transform the gap equation (13) to a more convenient expression. By setting $\Sigma = \uparrow$ ($\bar{\Sigma} = \downarrow$) and taking the summation of Λ , gap equation is given as

$$\begin{aligned} \lambda_E \Delta_{\uparrow\downarrow}^L(\mathbf{k}, \epsilon_n) &= \frac{\pi T}{(2\pi)^3} \sum_{\epsilon_m} \sum_M^{\text{FS}} \int_{\text{FSM}} \frac{d\mathbf{k}'_{\text{FSM}}}{v^M(\mathbf{k}')} \\ &\times \left\{ V_{\uparrow\downarrow\uparrow\downarrow}^{LM}(\mathbf{k}, \mathbf{k}', \epsilon_n - \epsilon_m) \frac{\Delta_{\uparrow\downarrow}^M(\mathbf{k}', \epsilon_m)}{|\epsilon_m|} \right. \\ &\quad \left. + V_{\uparrow\downarrow\downarrow\uparrow}^{LM}(\mathbf{k}, \mathbf{k}', \epsilon_n - \epsilon_m) \frac{\Delta_{\downarrow\uparrow}^M(\mathbf{k}', \epsilon_m)}{|\epsilon_m|} \right\} \end{aligned} \quad (22)$$

Since the relation $\Delta_{\uparrow\downarrow}^L(k) = -\Delta_{\downarrow\uparrow}^L(k)$ is satisfied, the gap equation for the singlet pairing state is given as

$$\begin{aligned} \lambda_E \Delta^L(\mathbf{k}, \epsilon_n) &= \frac{\pi T}{(2\pi)^3} \sum_{\epsilon_m} \sum_M^{\text{FS}} \int_{\text{FSM}} \frac{d\mathbf{k}'_{\text{FSM}}}{v^M(\mathbf{k}')} \\ &\times \left\{ V_{\uparrow\downarrow\uparrow\downarrow}^{LM}(\mathbf{k}, \mathbf{k}', \epsilon_n - \epsilon_m) \right. \\ &\quad \left. - V_{\uparrow\downarrow\downarrow\uparrow}^{LM}(\mathbf{k}, \mathbf{k}', \epsilon_n - \epsilon_m) \right\} \frac{\Delta^M(\mathbf{k}', \epsilon_m)}{|\epsilon_m|} \end{aligned} \quad (23)$$

where $\Delta^L(\mathbf{k}, \epsilon_n) \equiv \Delta_{\uparrow\downarrow}^L(\mathbf{k}, \epsilon_n)$.

III. SUPERCONDUCTING GAP FOR THE SOI-INDUCED BANDSTRUCTURE (I)

In this section, we numerically analyze the linearized Eliashberg equation, Eq. (23), on the basis of the SOI-induced bandstructure in Fig. 1 (a), which we call the “bandstructure (I)”. We show the obtained 3D gap function $\Delta^L(\theta, k_z)$ on the FS sheet L . Here, we divide the variables $\theta = [0, 2\pi]$ and $k_z = [-\pi, \pi]$ into 48 and 16 meshes, respectively, and use 512 Matsubara frequencies. The pairing interaction in Eq. (14) is given by the RPA.

A. Orbital-fluctuation-mediated s_{++} -wave state

We first discuss the s_{++} -wave state that is realized by orbital-fluctuations in the presence of the SOI: Figure 2 shows the obtained gap functions in the case of $g = 0.129$ eV and $U = 0$ ($\alpha_c = 0.98$) in the $k_z = \pi$ plane. The overall gap structure is essentially unchanged for $\alpha_c = 0.90 \sim 0.98$ [32]. As for the hole pockets, the gap functions on the h-FS2 composed of (d_{xz}, d_{yz}) orbitals are the largest in magnitude, while the gap on the h-FS3 composed of the d_{xy} orbital is the smallest. This result is quantitatively consistent with the experimental reports in Refs. [23, 34]: Experimental gap functions in Ref. [23] are shown by dots in Fig. 2. This result is also similar to our previous result without SOI, regardless of the presence or absence of the h-FS1 [32]. Here, we adjust the magnitude of gap functions given by solving the linearized gap equation.

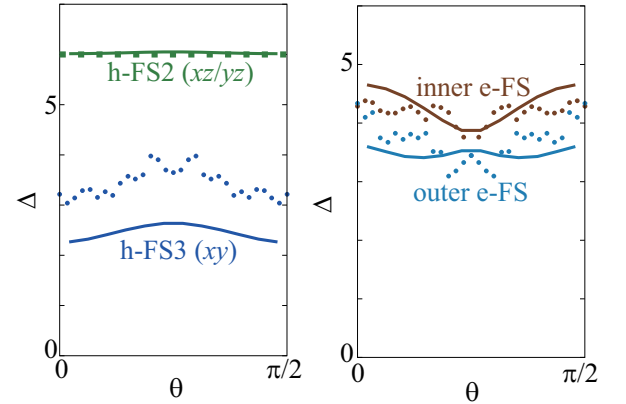


FIG. 2: (Color online) Obtained gap functions for $U = 0$ and $g = 0.129$ eV (s_{++} -wave state) in the $k_z = \pi$ -plane for the SOI-induced bandstructure (I). The dots represent the experimental data (in unit meV) given by the ARPES measurement in Ref. [23].

As for the electron pockets, the gap of the inner e-FS is larger than that of the outer e-FS, and has the local maxima at $\theta = 0$ and $\pi/2$ and the minima at $\theta = \pi/4$. This result is also consistent with the experimental data [23], and it is very similar to our previous result without SOI [32]. The obtained gap structure is almost indepen-

dent of k_z except for the three dimensional pocket h-FS2. These obtained results are essentially independent of the strength of g .

Next, we discuss the origin of the orbital and FS dependencies of the gap functions. For finite g , orbital fluctuations develop due to the good inter orbital nesting between h-FS2 (orbital 2,3) and e-FSs (orbital 4). For this reason, the maximum gap is realized on h-FS2 (Δ_2^h), and inner e-FS (Δ_{out}^e) at $\theta = 0$ and $\pi/2$. This result is not changed by considering the SOI. Therefore, the experimentally observed gap functions are understood based on the orbital-fluctuation theory.

B. Spin-fluctuation-mediated s_{\pm} -wave state

Next, we discuss the s_{\pm} -wave state realized by spin-fluctuations: Figure 3 shows the obtained gap structure in the case of $g = 0$ and $U = 0.439$ eV ($\alpha_s = 0.98$) in the $k_z = \pi$ plane. The overall gap structure is essentially unchanged for $\alpha_s = 0.90 \sim 0.98$ [32]. The gap function on each FS is almost independent of k_z . The obtained gap structure is essentially independent of the value of U . The obtained very small gap on the h-FS2 is totally opposite to the experimental data shown by dots.

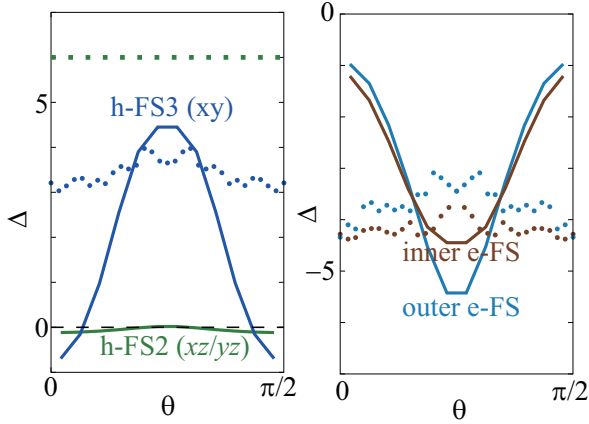


FIG. 3: (Color online) Obtained gap functions for $U = 0.439$ eV and $g = 0$ (s_{\pm} -wave state) in the $k_z = \pi$ -plane. The dots represent the experimental data (in unit meV) [23].

In addition, the obtained θ dependence of the gap on the e-FSs and h-FS3 is very different from the experimental data. Both the gap on the h-FS3 and e-FSs show the maximum values at $\theta = \pi/4$, since they are connected by the wave vector of spin-fluctuations $\mathbf{Q} \sim (\pi, 0), (0, \pi)$. Moreover, the gap function on h-FS3 has eight nodes, which is inconsistent with experiments. These results are very similar to our previous results without SOI [32]. Note that these eight nodes on the hole-FSs disappear in the case of $J/U = 0.4$ ($U' \equiv U - 2J = 0.2U$) in accordance with the previous study in Ref. [31].

C. Coexistence of orbital- and spin-fluctuations

Now, we discuss the superconducting state when the orbital- and spin-fluctuations coexist. In the previous results without SOI, coexistence of orbital- and spin-fluctuations leads to an exotic s -wave state, which has sign reversal within h-FSs: $\Delta_2^h \Delta_3^h < 0$ [32]. Here, we show that this "hole- s_{\pm} -state" is also realized in the presence of the SOI.

Figure 4 (a) shows the obtained gap functions in the case of $g = 0.125$ eV and $U = 0.200$ eV. The realized Stoner factors are $\alpha_c = 0.98$ and $\alpha_s = 0.45$. In this case, the orbital-fluctuations are much larger than the spin-fluctuations, and therefore we obtain the s_{++} -wave state. Except for h-FS3, the obtained gap structures are similar to those of the "pure s_{++} state" without U in Fig. 2. Due to the moderate spin-fluctuations on the d_{xy} orbital, the anisotropy of Δ_3^h is enlarged.

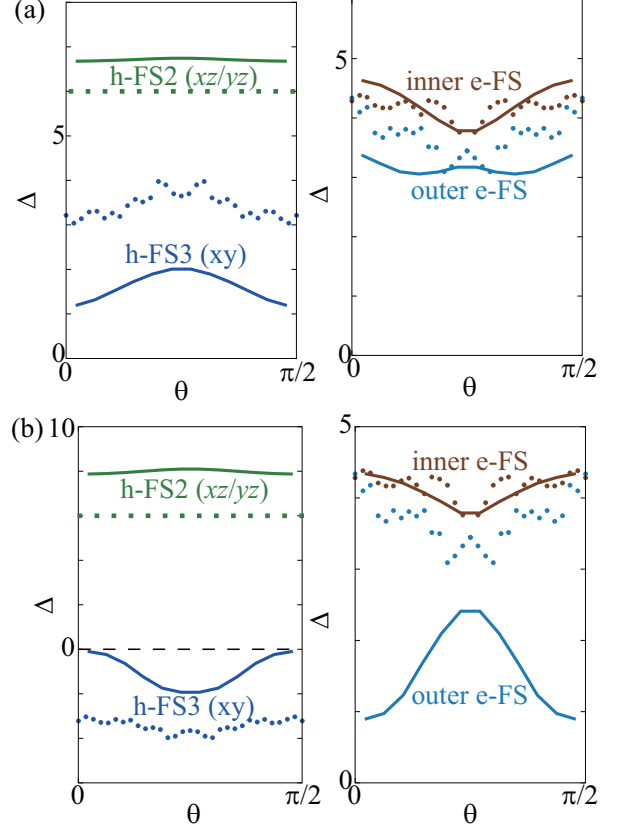


FIG. 4: (Color online) (a) Obtained gap functions for $U = 0.200$ eV and $g = 0.125$ (s_{++} -wave state) in the $k_z = \pi$ -plane. The dots represent the experimental data (in unit meV) [23]. (b) Obtained gap functions for $U = 0.380$ eV and $g = 0.122$ (hole- s_{\pm} -wave state) in the $k_z = \pi$ -plane.

If we increase the value of U further, we obtain the hole- s_{\pm} -state with the sign reversal within hole pockets. Figure 4 (b) shows the obtained gap functions in the case of $g = 0.122$ eV and $U = 0.380$ eV ($\alpha_c = 0.98$ and $\alpha_s = 0.85$). Here, only Δ_3^h is negative. In this hole- s_{\pm} -

wave state, the gap structure of Δ_2^h and that of the e-FSs (Δ^e) are qualitatively similar to those in the s_{++} -wave state in Fig. 2. On the other hand, Δ_3^h becomes very anisotropic, similarly to Δ_3^h in the s_{\pm} -wave state in Fig. 3.

In the hole- s_{\pm} -wave state given by the coexistence of orbital- and spin-fluctuations, $\Delta_2^h \cdot \Delta^e$ is positive due to orbital-fluctuations, whereas $\Delta_3^h \cdot \Delta^e$ is negative due to spin-fluctuations. Although the gap on the tiny hole-pocket in the hole- s_{\pm} -wave state takes the largest value, the overall gap structure is not consistent with ARPES measurement in Ref. [23]. The present hole- s_{\pm} -wave due to cooperation of orbital- and spin-fluctuation would be realized in other iron based superconductors. For instance, the hole- s_{\pm} -wave state is first discussed in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ based on the thermal conductivity and penetration depth measurements [44], in addition to the recent ARPES study [45].

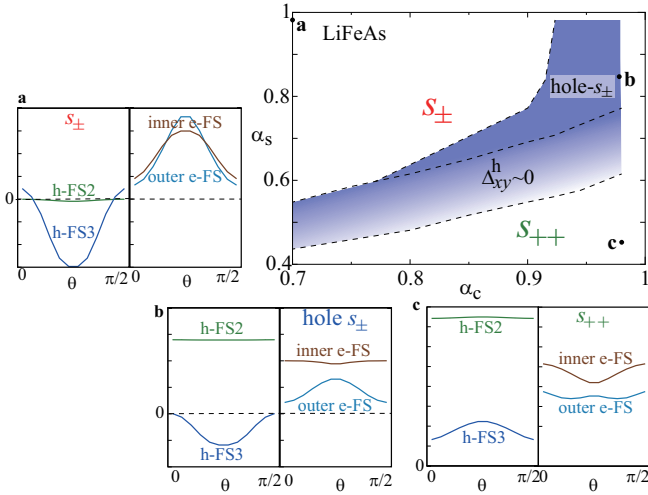


FIG. 5: (Color online) α_c - α_s phase diagram of the gap structure in LiFeAs obtained for the bandstructure (I) in Sec. III. Similar phase diagram is obtained for the bandstructure (II). The gap functions at each point **a** ~ **c** are shown.

Figure 5 shows the obtained α_c - α_s phase diagram of the gap structure in LiFeAs for $\lambda = 0.05$ eV (bandstructure (I)). As expected, the s_{\pm} -state (s_{++} -state) is realized for a wide region of $\alpha_c < \alpha_s$ ($\alpha_c > \alpha_s$). When both α_c and α_s are close to unity, we obtain the hole- s_{\pm} state gap in a wide region. The gap structure at each point **a** ~ **c** is shown in the figure. In the region " $\Delta_3^h \sim 0$ ", the obtained $\Delta_3^h(\theta)$ is very small and has line nodes, whereas the gaps on the other FSs are large and their signs are the same. Thus, nearly s_{++} -wave state is realized. Thus, various types of s -wave gap structures are realized due to the coexistence of orbital- and spin-fluctuations.

Finally, we comment that the hole- s_{\pm} state is also realized in the presence of strong repulsive pairing interaction within the h-FSs, as discussed in Refs. [49, 50]. The authors in Ref. [51] also discussed the hole- s_{\pm} state due to strong repulsive interaction between d_{xy} orbital

in h-FS3 and $d_{xz,yz}$ orbitals in e-FSs [51]. In the latter scenario, the gaps on e-FSs are nodal in the presence of the SOI. In contrast, the hole- s_{\pm} state in the present paper is realized by the cooperation between the "attractive interaction among (d_{xz}, d_{yz})- and d_{xy} -orbitals" and "repulsive interaction on the d_{xy} -orbital".

IV. ANOTHER SOI-INDUCED BANDSTRUCTURE AND GAP FUNCTION FOR LIFeAs

A. SOI-induced bandstructure (II)

Recently, the 3D bandstructure in LiFeAs had been carefully measured by ARPES study in Ref. [33]. The authors observed the strongly dispersing $d_{z^2} + d_{xy}$ -orbital band crossing the Fermi level along Γ -Z line, with the minimum at Z point. Although this dispersion is absent in bandstructure (I), it can be reproduced by lowering the d_{xy} -orbital level below the Fermi level at the Z point without changing d_{xy} -orbital h-FS3. To change the d_l -orbital level by ΔE_l ($l = xz, yz, xy, x^2 - y^2, z^2$) only around Z point, we introduce the additional intra-orbital hopping integrals shown in TABLE I. In this procedure, the d_l -level at M point in the unfolded Brillouin zone is unchanged. Figure 6 (a) shows the bandstructure for $\Delta E_{xy} = -0.6$ eV and $\Delta E_{xz} = \Delta E_{yz} = 0.01$ eV in the absence of the SOI. Due to large negative ΔE_{xy} , the strongly dispersing band along the Γ -Z line, shown by brown line, is reproduced. The energy of this dispersion is +0.21 eV (-0.51 eV) at Γ (Z) point. The Fermi level is set as zero. The FSs in the $k_y = 0$ plane are shown in Fig. 6 (b): After introducing large negative ΔE_{xy} , d_{xy} -level is lower than $d_{xz/yz}$ -level at Z-point, and the former level (latter level) corresponds to the energy of the hole-band for h-FS1 (h-FS2) at Z-point. As a result, the degeneracy of the hole-bands for h-FS1,2 at Z point is lifted, and only h-FS2 remains in Figs. 1 (a)-(b) whereas the hole-band for h-FS1 is below -0.1 eV. Thus, the number of the tiny 3D hole-pockets is reduced to one.

In the next stage, we introduce the SOI ($\lambda = 0.05$ eV): The realized SOI-induced bandstructure is shown in Fig. 6 (c), which we call the "SOI-induced bandstructure (II)". Due to the SOI, the degenerated $d_{xz/yz}$ -orbital hole-bands along the Γ -Z line, is split into two bands, as shown by purple lines in Fig. 6 (c). As a result, the size of h-FS2 is reduced as explained in Ref. [33] in detail. (In contrast, the higher band forms the tiny hole-pocket in the SOI-induced bandstructure (I) in Sec. III.)

The obtained SOI-induced FSs in the $k_y = 0$ plane are shown in Fig. 6 (d): In addition to the tiny and large hole-pockets h-FS2 and h-FS3, there are fine h-FSs due to the Dirac dispersion at $|k_z| \sim 0.5\pi$ on the k_z -line. The overall bandstructure near the Fermi level as well as the SOI-induced change in the topology of the FSs reported in Ref. [33] are qualitatively reproduced in the present bandstructure (II).

TABLE I: Additional hopping integrals $\Delta t_{l,l}(\mathbf{R})$ to change the d_l -orbital level by ΔE_l only around the Z point, without changing the d_l -level at M point in the unfolded Brillouin zone. $s_l = +1$ (-1) for $l = z^2, xy, x^2 - y^2$ (xz, yz).

\mathbf{R}	[0,0,0]	[1,0,0]	[1,1,0]	[2,0,0]	[2,1,0]
$\Delta t_{l,l}(\mathbf{R})$	$(1/16)\Delta E_l$	$-(3s_l/64)\Delta E_l$	$(1/32)\Delta E_l$	$(1/64)\Delta E_l$	$-(s_l/128)\Delta E_l$
\mathbf{R}	[0,0,1]	[1,0,1]	[1,1,1]	[2,0,1]	[2,1,1]
$\Delta t_{l,l}(\mathbf{R})$	$-(1/32)\Delta E_l$	$(3s_l/128)\Delta E_l$	$-(1/64)\Delta E_l$	$-(1/128)\Delta E_l$	$(s_l/256)\Delta E_l$

B. Superconducting gap for the bandstructure (II)

Next, we analyze the linearized Eliashberg equation, Eq. (23), for the SOI-induced bandstructure (II): The FSs in the $k_z = \pi$ plane are shown in Fig. 7 (a). We stress that the orbital character of h-FS2 in Fig. 7 (a) is essentially the same as that of h-FS2 in Fig. 1 (c). This fact indicates that similar gap structures are obtained based on both bandstructures (I) and (II).

The pairing interaction is given by Eqs. (14) and (15) in the present model with ΔE_l . We first discuss the s_{++} -wave state realized by orbital-fluctuations: Figure 7 (b) shows the obtained gap functions in the case of $g = 0.127$ eV and $U = 0$ ($\alpha_c = 0.98$) in the $k_z = \pi$ plane. As for the hole pockets, the gap functions on the h-FS2 composed of (d_{xz}, d_{yz}) orbitals are the largest, while the gap on the h-FS3 composed of the d_{xy} orbital is the smallest. These results are quantitatively consistent with the experimental data [23] shown by dots. As for the electron pockets, the gap of inner e-FS is larger than that of outer e-FS, and their θ dependences are qualitatively consistent with experimental data in Ref. [23]. The obtained overall gap structure is similar to that for the bandstructure (I) shown in Sec. III A, regardless of significantly different bandstructures.

Next, we discuss the s_{\pm} -wave state realized by spin-fluctuations: Figure 7 (c) shows the obtained gap structure in the case of $U = 0.434$ eV and $g = 0$ ($\alpha_s = 0.98$) in the $k_z = \pi$ plane. The obtained very small gap functions on the h-FS2 is opposite to the experimental data [23] shown by dots. In addition, the obtained θ dependences of the gaps on the e-FSs and h-FS3 are very different from the experimental data: Both the gaps on the h-FS3 and e-FSs show the maximum values at $\theta = \pi/4$ since they are connected by the wave vector of spin-fluctuations $\mathbf{Q} \sim (\pi, 0), (0, \pi)$. These results are similar to those for the bandstructure (I) shown in Sec. III B.

V. SUMMARY

In this paper, we studied the pairing mechanism in LiFeAs by taking the SOI into account. For this purpose, we constructed possible two realistic 3D tight-binding models with SOI, according the experimental band structures [33, 34]. In both SOI-induced bandstructures, one of the 3D tiny hole-pockets around the Z point disappears due to the SOI. The orbital character of the realized tiny

single hole-pocket is same in both models. Experimentally, the superconducting gap takes the largest value on the tiny single hole-pocket.

Next, we analyzed the 3D gap structure in order to determine the correct pairing mechanism. In the orbital-fluctuation scenario for the s_{++} -wave state, the pairing interaction is given by the orbital-fluctuations driven by the inter-orbital nesting between $(d_{xz/yz}, d_{xy})$ orbitals. The obtained gap function on the tiny hole-pocket is the largest, by reflecting the better inter-orbital nesting between the tiny hole-pocket and the electron-pockets. The obtained numerical results shown in Fig. 2 and Fig. 7 (b) are quantitatively consistent with the experimental gap structure in LiFeAs. In contrast, in the spin-fluctuation scenario for the s_{\pm} -wave state, the obtained gap function on the tiny hole-pocket is the smallest, by reflecting the bad intra-orbital nesting between the tiny hole-pocket and the electron-pockets. Thus, the experimental gap structure in LiFeAs is unable to be explained by this scenario, as demonstrated in Fig. 3 and in Fig. 7 (c).

Therefore, it is concluded that the inter-orbital $(d_{xz/yz}-d_{xy})$ interactions, which are given by orbital-fluctuations, cause the superconductivity in LiFeAs. The present study reinforces the results obtained in our previous study for LiFeAs without the SOI [32]. When both the orbital- and spin-fluctuations develop comparably, we frequently obtain the “hole- s_{\pm} -state”, in which the gap structure has the “sign reversal between h-FSs”. This interesting gap structure had been discussed in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ based on the thermal conductivity and penetration depth measurement [44]. In Appendix A, we show that the hole- s_{\pm} -wave gap structure can be obtained in the model of Ba122 system.

Acknowledgments

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Appendix A: Hole- s_{\pm} -wave state due to coexistence of orbital- and spin-fluctuations in Ba122 systems

In this appendix, we show that the hole- s_{\pm} -wave can be realized in Ba122 compounds under the coexistence of orbital- and spin-fluctuations. To construct the

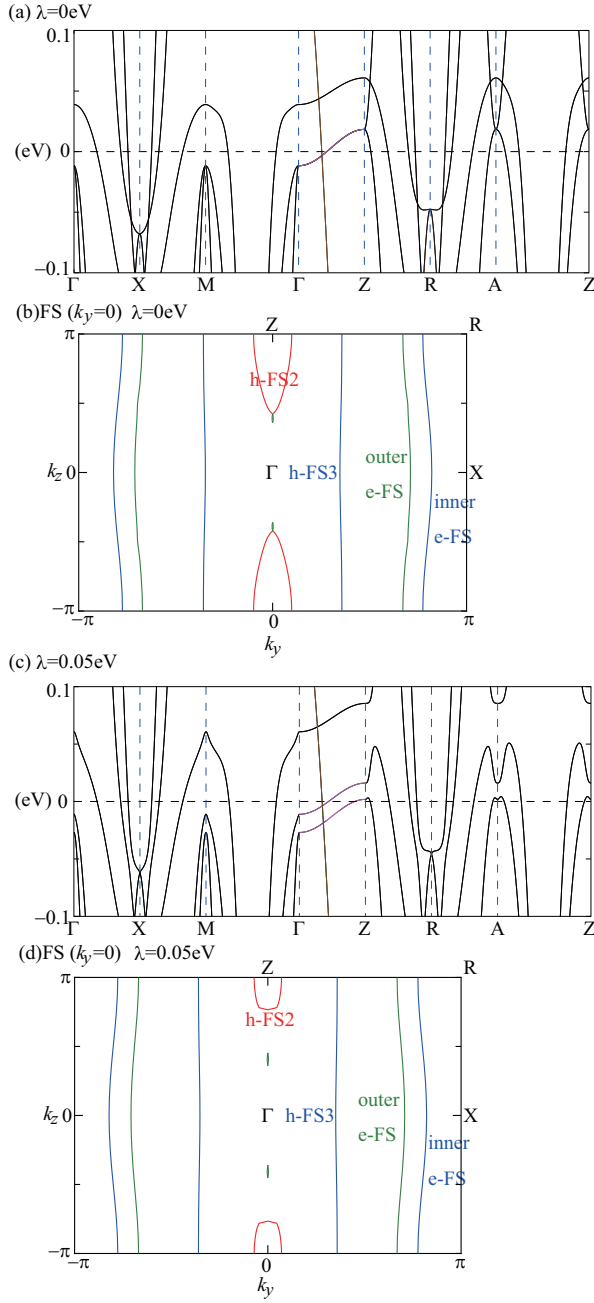


FIG. 6: (Color online) (a) The band structure of the ten-orbital model for $\Delta E_{xy} = -0.6$ eV and $\Delta E_{xz} = \Delta E_{yz} = 0.01$ eV without the SOI ($\lambda = 0$) and (b) the FSs in the $k_y = 0$ plane for $\lambda = 0$. Due to large negative ΔE_{xy} , h-FS1 around Z point disappears. (c) The “SOI-induced bandstructure (II)” with $\lambda = 0.05$ eV, and (d) the corresponding FSs in the $k_y = 0$ plane. The size of h-FS2 is reduced by introducing the SOI.

bandstructure, we perform a local-density-approximation (LDA) band calculation for BaFe_2As_2 using the WIEN2K code based on the experimental crystal structure. Next, we derive the ten-orbital tight-binding model using the WANNIER90 code and the WIEN2WANNIER interface [52]. However, the d_{xy} -orbital h-FS given by the

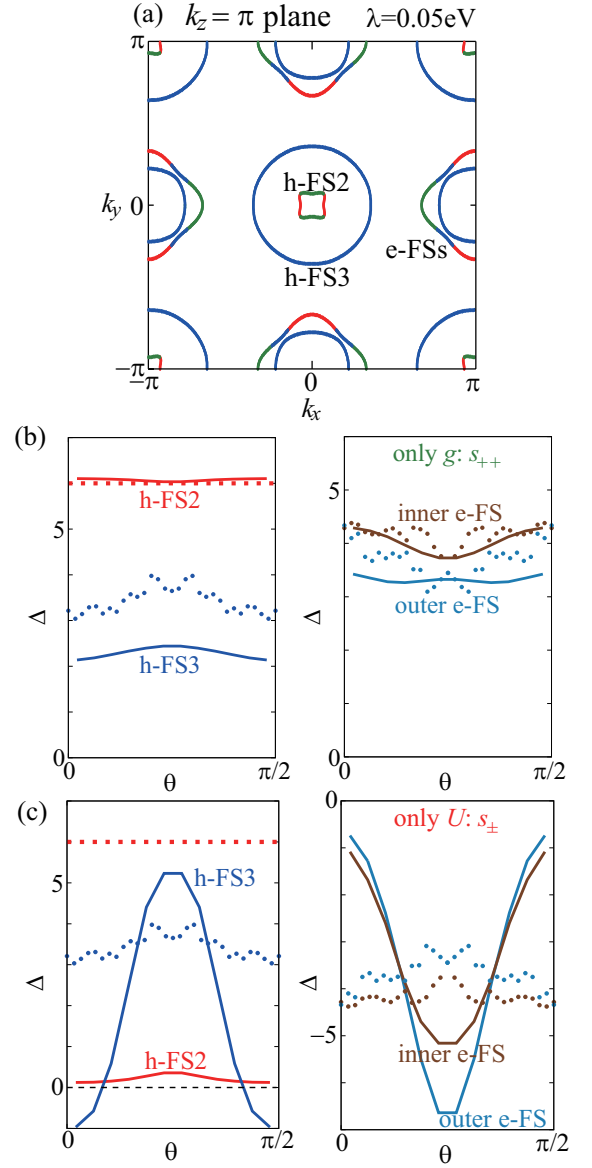


FIG. 7: (Color online) (a) FSs in the bandstructure (II) with $\lambda = 0.05$ eV in the $k_z = \pi$ plane. (b)-(c) The obtained gap functions in bandstructure (II) in the $k_z = \pi$ -plane for (b) $g = 0.127$ eV and $U = 0$ (s_{++} -wave state), and (c) $U = 0.434$ eV and $g = 0$ (s_{\pm} -wave state). The dots represent the experimental data given by ARPES study (in unit meV) [23].

LDA is much smaller than the reports by ARPES studies. In order to magnify its size, we introduce the additional dependent potential Γ -Z line: $H_{\text{kin}} = H^0 + \sum_{l\sigma\mathbf{k}} e_{l\frac{1}{2}}(\cos k_x \cos k_y + 1)c_{l\sigma}^\dagger(\mathbf{k})c_{l\sigma}(\mathbf{k})$, where e_l is the energy shift of the orbital l in the k_z -axis. We put $e_{xy} = 0.15$ eV, and the others are 0. The obtained FSs in the $k_z = 0$ plane are shown in Fig. 8 (a). Near the Γ point, the d_{xy} -orbital h-FS is largest of all h-FSs. Here, we neglect the SOI since the SOI-induced modification of the FSs is very small in Ba122 systems.

Next, we solve the Eliashberg equation by introduc-

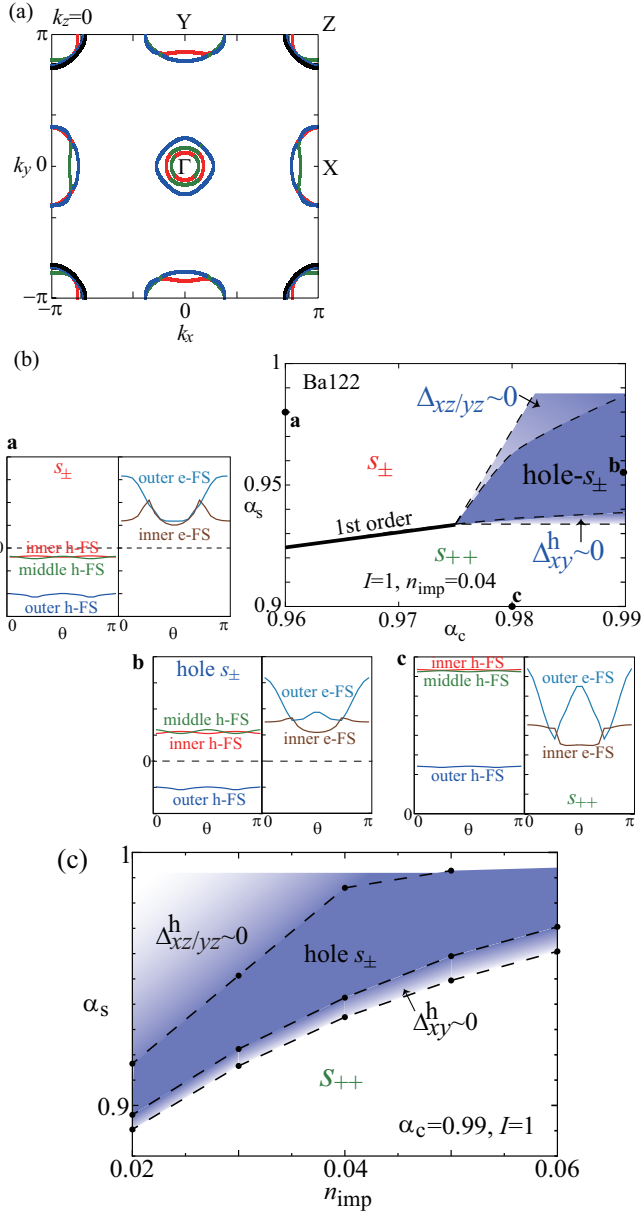


FIG. 8: (Color online) (a) The FSs of the three dimensional model for BaFe_2As_2 in the $k_z = 0$ plane. The green, red, blue, and black lines correspond to xz , yz , xy , and z^2 orbitals, respectively. (b) The obtained α_c - α_s phase diagram. The gap structure at each point $\mathbf{a} \sim \mathbf{c}$ is shown in the figure. The horizontal axis is $\theta = \tan(\bar{k}_y/\bar{k}_x)$, where (\bar{k}_x, \bar{k}_y) is the momentum on the FS with the origin at the center of each pocket. (c) n_{imp} - α_s phase diagram in the superconducting state. The hole- s_{\pm} state is realized for wide region in the phase diagram.

ing dilute concentrations of impurities ($I = 1$ eV and $n_{\text{imp}} = 0.04$) by applying the T -matrix approximation [18] to make the obtained gap structures smoother. Figure 8 (b) shows the obtained α_c - α_s diagram of the superconducting state. The gap functions on the FSs (in the $k_z = 0$ plane) at each point $\mathbf{a} \sim \mathbf{c}$ are shown in the

figure. The obtained phase diagram is qualitatively similar to that for LiFeAs shown in Fig. 5. Note that the gap structure in the region " $\Delta_{xy}^h \sim 0$ ($\Delta_{xz/yz}^h \sim 0$)" is similar to the s_{++} (s_{\pm})-wave state.

In the hole- s_{\pm} -wave state at point \mathbf{b} , only the gap function on the outer h-FS (d_{xy} -orbital) is sign-reversed. We stress that the gap on the inner e-FS takes the minimum at $\theta = \pi/2$ in the $k_z = 0$ -plane, which is consistent with ARPES measurement for $\text{BaFe}_2(\text{As,P})_2$ [20]. This hole- s_{\pm} -wave state is changed to the s_{++} -wave state (point \mathbf{c}) by increasing the impurities. During this change, large impurity-induced DOS appears at the Fermi level due to the outer h-FS, which may correspond to the experimental disorder-induced gap structure in $\text{BaFe}_2(\text{As,P})_2$ reported in Ref. [54].

Figure 8 (c) shows the n_{imp} - α_s phase diagram in the superconducting state, where n_{imp} is concentration of the non-magnetic impurity ($I = 1\text{eV}$). The hole- s_{\pm} state is realized for wide region in the phase diagram, reflecting the fact that the hole- s_{\pm} state is realized by the "cooperation" between orbital- and spin-fluctuations. That is, the "intra-orbital repulsive interaction by spin-fluctuations" mainly works between outer h-FS and e-FSs, whereas the "inter-orbital attractive interaction by orbital-fluctuations" mainly works between inner and middle h-FSs ($d_{xz/yz}$ -orbitals) and e-FSs. In the SC- VC_{Σ} theory, both s_{++} and hole- s_{\pm} states are realized in the case of $\alpha_c \sim \alpha_s$ even for $n_{\text{imp}} = 0$, since the electron-orbital coupling constant ($\sim U'$) is enlarged by the vertex corrections for the gap equation (Δ -VC) [53]. A crossover from hole- s_{\pm} state to s_{++} state may be realized in $\text{BaFe}_2(\text{As,P})_2$ by electron irradiation in Ref. [54].

Appendix B: Spin susceptibility in the presence of the SOI

In the main text, we studied the superconducting state in LiFeAs in the presence of the SOI. However, we neglected the SOI in the spin and charge susceptibilities in the pairing interactions $\hat{V}^s(\mathbf{q})$ and $\hat{V}^c(\mathbf{q})$, respectively, given in Eq. (16). This approximation is justified if the relation $\chi_x^s(\mathbf{q}) \approx \chi_y^s(\mathbf{q}) \approx \chi_z^s(\mathbf{q})$ is satisfied, although this relation is not exact in the presence of the SOI. In order to verify this relation, here we calculate the spin susceptibilities in the presence of the SOI using the RPA.

In the presence of the SOI, the spin is not conserved. Therefore, the Green function is given by the following 20×20 matrix form, $\hat{G}(\mathbf{k}) = (i\epsilon_n + \mu - \hat{h}(\mathbf{k}))^{-1}$, where $\hat{h}(\mathbf{k})$ is given in Eq. (9). Then, the irreducible susceptibility is $\hat{\chi}_{l\sigma, l'\sigma'; m\rho, m'\rho'}^0(\mathbf{q}) = -T \sum_{n, \mathbf{k}} G_{l\sigma, m\rho}(\mathbf{q} + \mathbf{k}) G_{m'\rho', l'\sigma'}(\mathbf{k})$.

In the RPA, the susceptibility in the presence of the SOI is given by the following $20^2 \times 20^2$ matrix form:

$$\hat{\chi}(\mathbf{q}) = (1 - \hat{\chi}^0(\mathbf{q})\hat{\Gamma})^{-1}\hat{\chi}^0(\mathbf{q}), \quad (\text{B1})$$

where $\hat{\Gamma}$ is the Coulomb interaction in the matrix form: $\Gamma_{ll', l''\sigma'; m\rho, m'\rho'} = \Gamma_{ll', mm'}^c \delta_{\sigma, \rho'} \delta_{\sigma', \rho} + \Gamma_{ll', mm'}^s \boldsymbol{\sigma}_{\sigma, \rho'} \cdot \boldsymbol{\sigma}_{\sigma', \rho}$.

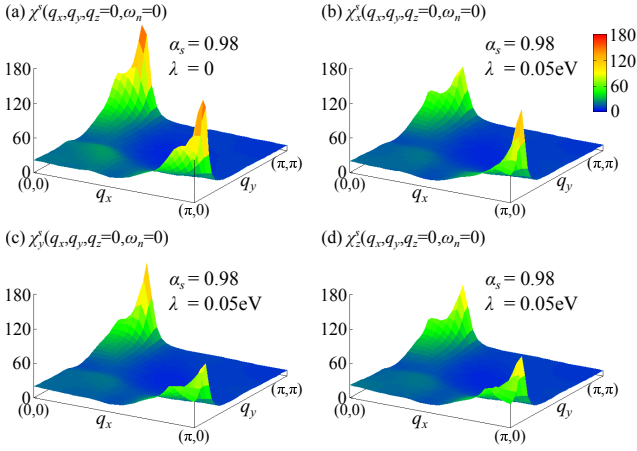


FIG. 9: (Color online) Spin susceptibilities in the case of $\alpha_S = 0.98$. (a) $\chi^s(\mathbf{q})$ for in the absence of the SOI. (b) $\chi_x^s(\mathbf{q})$, (c) $\chi_y^s(\mathbf{q})$ and (d) $\chi_z^s(\mathbf{q})$ in the presence of the SOI ($\lambda = 0.05$ eV).

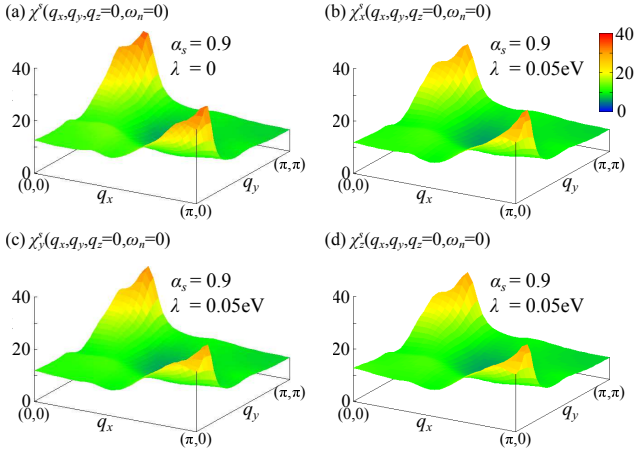


FIG. 10: (Color online) Spin susceptibilities in the case of $\alpha_S = 0.90$. (a) $\chi^s(\mathbf{q})$ for in the absence of the SOI. (b) $\chi_x^s(\mathbf{q})$, (c) $\chi_y^s(\mathbf{q})$ and (d) $\chi_z^s(\mathbf{q})$ in the presence of the SOI ($\lambda = 0.05$ eV).

Then, the spin susceptibility in the μ -direction is

$$\chi_\mu^s(\mathbf{q}) = \frac{1}{4} \sum_{l,m,\sigma,\sigma',\rho,\rho'} \chi_{l\sigma,l\sigma';m\rho,m\rho'}(\mathbf{q}) \sigma_{\sigma',\sigma}^\mu \sigma_{\rho,\rho'}^\mu \quad (\text{B2})$$

where $\mu = x, y, z$.

In Fig. 9, we show the numerical results in the vicinity of the magnetic critical point, at which the spin Stoner factor is $\alpha_S = 0.98$. Figure 9 shows the obtained spin susceptibilities for $\lambda = 0$ ($U = 0.439$ eV) in (a) and for $\lambda = 0.05$ eV ($U = 0.446$ eV) in (b)-(d), respectively. In the case of $\alpha_S = 0.98$, the effect of the SOI on the spin susceptibility is not small. In contrast, we verified that the effect of the SOI on the charge susceptibility $\chi_{l,l';m,m'}^c(\mathbf{q}) = \sum_{\sigma,\rho} \chi_{l\sigma,l\sigma';m\rho,m\rho'}(\mathbf{q})$ is very small even for $\alpha_C = 0.98$.

In Fig. 10, we show the numerical results when the spin fluctuations are moderate ($\alpha_S = 0.90$), which is consistent with the weak spin fluctuations observed in LiFeAs. Figure 9 shows the obtained spin susceptibilities for $\lambda = 0$ ($U = 0.403$ eV) in (a) and for $\lambda = 0.05$ eV ($U = 0.409$ eV) in (b)-(d), respectively. In the case of $\alpha_S = 0.90$, the relation $\chi_x^s(\mathbf{q}) \approx \chi_y^s(\mathbf{q}) \approx \chi_z^s(\mathbf{q})$ is approximately satisfied, so the rotational symmetry of the spin susceptibility is almost satisfied. Thus, the effect of the SOI on the pairing interaction is expected to be negligible.

Experimentally, LiFeAs is not close to the magnetic critical point because the observed spin fluctuations are small, so the relation $\alpha_S \sim 0.9$ is expected in LiFeAs. We have verified that the gap structure in the s_{\pm} -state shown in Fig. 3 is essentially unchanged even for $\alpha_S = 0.90$, similarly to our previous study in Ref. [32]. Therefore, the effect of the SOI on the pairing interaction $\hat{V}^{s,c}(\mathbf{q})$ in LiFeAs is negligible, so the obtained results in the main text are essentially unchanged even if the SOI is included in the pairing interaction.

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